Piezoelectric Response of Domains in Ferroelectrics

Avadh Saxena and Turab Lookman, T-11; and Rajeev Ahluwalia, Institute for Materials Research & Engineering, Singapore; and W. Cao, Pennsylvania State University

Ferroelectrics are the best piezoelectric materials that can convert electrical energy into mechanical energy and vice versa. This electromechanical property arises due to the coupling of the spontaneous polarization with lattice strain. Many devices such as ultrasonic transducers and piezoelectric actuators make use of this property. Recently, there has been considerable interest in this field due to the observation of a giant piezoelectric response when the applied field is along a nonpolar direction. It is believed that this "superpiezoelectric" response is due to the symmetry change caused by a rotation of the polarization towards the direction of the applied field. Domain configurations produced by the nonpolar direction field are termed *engineered domains*. It is important to understand to what extent the large response is influenced by the presence of domain walls in the engineered configuration. Recent studies have shown that the low field piezoelectric response of the engineered domain configuration of $0.67Pb(Mg_{1/3}Nb_{2/3})O_3 - 0.33PbTiO_3$ is very close to that of single domain data. However, for large fields, the presence of domain walls may influence the piezoelectric response. We have theoretically studied the effect of domain walls on the piezoelectric properties of engineered domains as a function of the applied field over a range of electric field values.

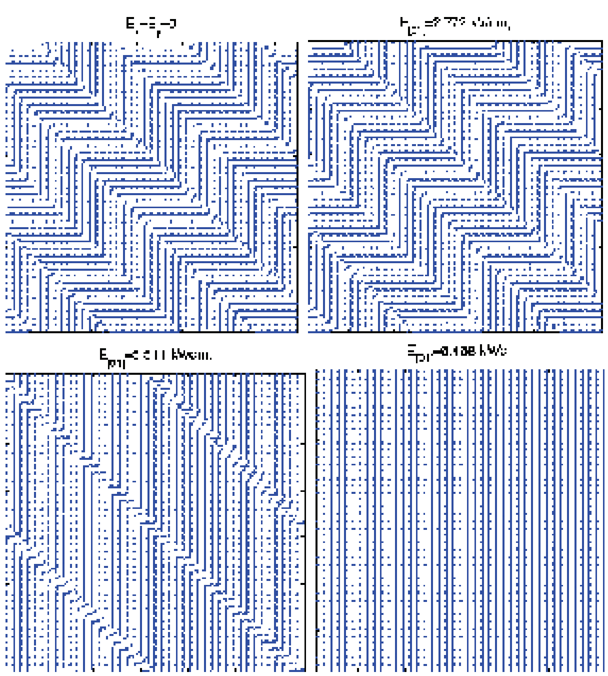

Fig. 1.
The top left panel of the figure shows the prepared zero-field multidomain state. To simulate the effect of an external electric field, the evolution equations are solved with a varying field for two cases: (i) the field applied along the polar [01] direction, and (ii) the field is applied along the nonpolar [11] direction. The figure depicts the evolution of domains for the field along the polar direction.

In previous studies, the electromechanical response of ferroelectrics poled along nonpolar directions has been studied theoretically using first-principle calculations. A continuum Landau theory describing a single domain or homogeneous state has also been used to study electromechanical properties of $BaTiO_3$ as a function of temperature and electric field direction. Although such calculations provide valuable insights into the physics of the polarization-strain coupling, they do not describe inhomogeneities due to domains and domain walls. We have used a continuum time-dependent Ginzburg-Landau framework that incorporates long-range elastic and electrostatic interactions that are crucial to describe multidomain states and domain walls. To illustrate the underlying principles, we restrict ourselves to a 2-D ferroelectric system. However, we choose free energy parameters relevant for $BaTiO_3$. We have also studied the behavior of domain patterns under applied electric field and investigated how the microstructural evolution influences the average electromechanical response of ferroelectric materials.

In the top left panel of Fig. 1 we show the prepared zero-field multidomain state. To simulate the effect of an external electric field, the evolution equations are solved with a varying field for two cases: (i) the field applied along the polar [01] direction, and (ii) the field is applied along the nonpolar [11] direction. The evolution of domains for the field along the polar direction is depicted in Fig. 1. The corresponding (longitudinal) piezoelectric constant d_{33}, derived from the relevant polarization and strain, is shown in Fig. 2 for three different domain sizes (4.5 nm, 11.3 nm, 22.6 nm from left to right, respectively). The inset shows the low-field behavior. We find that the electromechanical response is highly orientation- and microstructure-dependent. When the field is applied along one of the polar directions, domain switching results in higher strains compared to the single domain state. We have also studied the domain evolution, polarization, strain, and the associated piezoelectric constant for electric fields applied along a nonpolar direction. In this case the domain walls serve as nucleation sites for a field-induced structural phase transition.

To summarize, we have used a Ginzburg-Landau formalism to study the electromechanical properties of domain-engineered ferroelectrics. The model calculation incorporates nonlocal elastic and electrostatic effects. The simulations demonstrate that domain walls do not significantly influence the piezoelectric response at small fields. However, at high fields, domain walls act as nuclei for field-induced structural phase transitions and consequently the transition occurs at lower field values for the engineered state compared to the corresponding single domain states. The high-field piezoelectric response is also enhanced due to the nucleating action of the domain walls near the field-induced transition. The low field behavior is consistent with the reported results on $0.67Pb(Mg_{1/3}Nb_{2/3})O_3 - 0.33PbTiO3$. Further experiments would test the large field predictions of our calculations. We also demonstrated the effect of the underlying domain microstructure on electromechanical properties of multidomain ferroelectrics. To account for nonlocal elastic and electrostatic effects, two additional parameters that measure the strength of the long-range interactions were introduced. Our calculations show that these long-range parameters are essential to describe multidomain states. In this work, we have made simple choices for these parameters. In principle, these parameters should be measured for a given experimental multidomain state for a complete characterization of the material.

For more information contact Avadh Saxena at avadh@lanl.gov.

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[2] R. Ahluwalia, et al., *Phys. Rev. B* **72**, 014112 (2005).

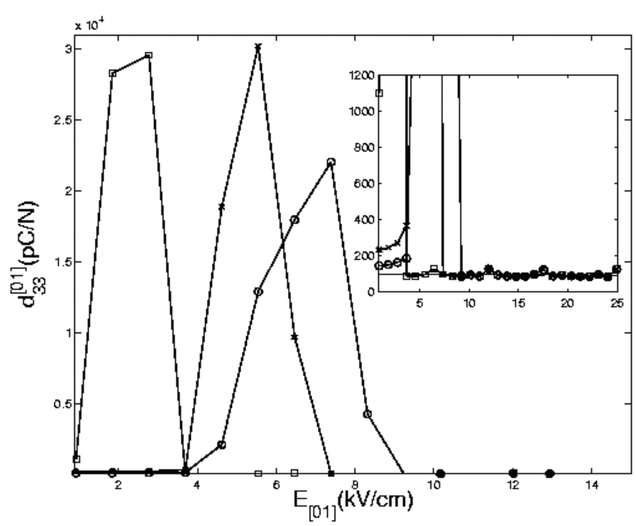

Fig. 2. *The longitudinal piezoelectric constant d_{33}, derived from the relevant polarization and strain for three different domain sizes (4.5 nm, 11.3 nm, 22.6 nm from left to right, respectively). The inset shows the low-field behavior.*